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Effect of plasmonic Au nanoparticles on IR luminescence of Ag₂S quantum dots

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Abstract

Luminescent manifestations of the interaction of Ag_2S quantum dots (QDs) with Au nanorods (NRs) depending on the overlap degree of the corresponding luminescence bands and plasmon resonance peaks have been experimentally established. Under spectral resonance conditions, the possibility of controlling the intensity of QDs luminescence by changing the interaction with Au NRs by varying the distance between the components of the plasmon-exciton mixture has been demonstrated. In turn it determines the influence of the near-field of metal nanoparticles on photoprocesses in Ag_2S QDs.

The detuning of the spectral resonance due to the change in the Au QDs length leads to the asymmetry of the spectral contour of the Ag₂S QDs luminescence band, which may be due to the manifestation of the Fano effect during plasmon-exciton interaction, taking into account the inhomogeneous broadening of the corresponding bands.

Keywords: Quantum dot, Plasmonic nanoparticles, Plasmon resonance, IR luminescence, Plasmon-exciton interaction

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1. Introduction

Semiconductor colloidal quantum dots (QDs) are relevant objects of research due to their sizedependent spectral-luminescent properties, which ensure control of the the corresponding peaks by varying the size without changing the chemical composition of the nanocrystals [1-9]. Such unique spectral properties of colloidal QDs make them promising materials for a wide range of practical applications in modern photonics, including optoelectronics [1, 2, 7–9], luminescence and biosensors [2, 5, 6], photocatalysis [1–4], and others.

Additional opportunities for controlling the spectral-luminescent properties of QDs are provided by the use of exciton-exciton and plasmon-exciton interaction effects [10-21]. Plasmon-exciton interaction, realized during the association of colloidal QDs with plasmonic nanoparticles, leads to striking effects [12-21]. The physical mechanism of plasmon-exciton effects is associated with the mutual near-field interaction of QDs and NPs, in which the NPs acts as both a source of strong polarization of the environment and a nano-resonator concentrating the field energy [13, 14, 17]. A lot of experimental studies, implemented primarily within the framework of single-object spectroscopy techniques, demonstrate an increase in the intensity of QDs luminescence with a simultaneous decrease in its duration in the presence of plasmonic NPs, which is interpreted as a manifestation of the Purcell effect [12, 13, 18–21]. Depending on the magnitude of the mutual distance between the components of the plasmon-exciton structure and spectral tuning of resonances in the luminescence and extinction spectra of plasmonic NPs, the occurrence of the effect of quantum interference (Fano effect) is probable, as well as the splitting of the luminescence spectra under conditions of strong plasmon-exciton coupling (Rabi splitting) [13, 15–17]. The development of applications related to the use of plasmon-exciton effects in luminescent sensors requires an understanding of the interactions that arise between NPs and QDs, as well as their manifestation in the luminescence of ensembles of QDs [18, 19, 22–24]. The dispersion of QDs in size in the ensemble suggests a noticeable broadening of their luminescence band, as well as offset of the

spectral resonance, which determines the effects of interaction in the resulting spectral properties of plasmon-exciton nanostructures [18, 19, 22-24]. In addition, the necessary conditions for observing the effects of plasmon-exciton interaction (spectral resonance of the extinction peak of NPs and the luminescence band of QDs, the distance between the components) suggest the presence of other, accompanying interaction processes, in particular, non-radiative energy transfer between QDs and NPs [25], as well as photoinduced charge transfer [26, 27]. In this regard, the manifestation of plasmon-exciton interaction effects in the luminescent properties of colloidal ensembles of QDs may differ significantly from the manifestation of these effects for a single emitter.

This study is devoted to the experimental establishment of the influence of Au plasmonic nanorods (NRs) on the IR trap state luminescence of Ag₂S ODs ensembles depending on the degree of overlap of the plasmon resonance peak and luminescence band.

2. Experimental

Synthesis of colloidal Ag_2S QDs was carried out in ethylene glycol. The 2-mercaptopropionic acid (2-MPA) molecules were used as a passivating ligand [28]. The synthesis method involved mixing $AgNO_3$ precursors and 2-MPA in a molar ratio of 1:2 in 30 ml of ethylene glycol. The control of particle size and position of the luminescence peak within this approach was achieved by exposing a colloidal solution of Ag_2S QDs by radiation with a wavelength of 405 nm (100 mW) for 20 hours. The removal of reaction by-products, colloidal Ag_2S QDs were precipitated by centrifugation and re-dissolution in ethylene glycol.

Plasmonic Au NRs were obtained by aqueous synthesis using cetyltrimethylammonium bromide (CTAB) molecules, which form cylindrical micelles, thereby defining anisotropic conditions for NRs growth [18]. The length of the Au NRs was controlled by adding $AgNO_3$ to the reaction solution. The changes in the length of the Au NRs allows to control the position of the extinction peak of the longitudinal plasmon resonance of the Au NRs, ensuring spectral resonance with the luminescence band of the Ag₂S QDs.

In the Ag_2SQDs colloidal solution Au NRs were introduced in a concentration ratio ~ 10⁴:1 pcs. for the formation of plasmon-exciton mixtures. Solutions containing Ag_2SQDs and Au NRs separately in concentrations equivalent to those introduced during the formation of their mixtures served as comparison samples in the study of spectral-luminescent properties of mixtures of Ag_2SQDs and Au NRs.

Structural data of Ag, S QDs and Au NRs were determined using a Libra 120 transmission electron microscope (TEM) (CarlZeiss, Germany) and a JEOL 2000FX high-resolution TEM (JEOL Ltd., Japan). The absorption properties were studied using a USB2000+ spectrometer (OceanOptics, USA) with a USB-DT radiation source (OceanOptics, USA). Luminescence spectra and luminescence decay of Ag₂S QDs were studied using USB2000+ and TimeHarp~260 system for time correlated photon counting (PicoQuant Germany) with a PMC-100-20 photomultiplier tube (Becker&Hickl Germany) with a time resolution of 0.2 ns. A diode laser LD PLTB450 (Osram, Germany) with a wavelength of 445 nm (200 mW) was used to stimulate the luminescence.

3. Results and discussion

3.1. Structural properties

The analysis of TEM images of Ag_2S ODs showed the formation of individual nanocrystals with an average size of 2.8 ± 0.5 nm with an ensemble dispersion of ~30%, which was due to the chosen approach of colloidal synthesis in an aqueous solution (Fig. 1a).

According to TEM images, the formation of Au NRs with average length values of 30 ± 5 nm and 35 ± 5 nm and a diameter of 9 ± 2 nm (Fig. 1b) and Au NRs with an average length of 45 ± 5 nm and a diameter of 9 ± 2 nm was established (Fig. 1c). The dispersion of the Au NRs in the ensemble did not exceed 30% (Fig. 1b, c).

High-resolution TEM data (Fig. 1d) showed cluster of spherical nanoparticles near cylindrical nanoparticles. The interplanar distance of ~ 0.251 nm of spherical nanoparticles corresponded to the crystallographic plane (022) of the monoclinic crystal lattice of Ag₂S, and the interplanar distance of 0.237 nm of cylindrical nanoparticles corresponded to the crystallographic plane (111) of the face-centred cubic crystal lattice of Au (Fig. 2d).



Fig. 1. TEM image of $Ag_2SQDs - (a)$. TEM image of Au NRs with average length of 30 nm and 35 nm – (b). TEM image of Au NRs with average length of 45 nm – (c) High resolution TEM image of Ag_2SQDs and Au NRS – (d). Optical absorption (1) and luminescence (2) spectrum of Ag_2SQDs , extinction spectra of Au NRs with average length of 30 nm (3), 35 nm (4) and 45 nm (5) – (e)



Fig. 2. Extinction spectrum of Au NRs with an average length of 45 nm (1), luminescence spectra of free Ag_2S QDs (2), in the presence of plasmonic Au NRs (3) and in the presence of plasmonic Au NRs and polymer (4) – (a). Extinction spectrum of Au NRs with an average length of 30 nm (1) and 35 nm (2), luminescence spectra of free Ag_2S QDs (3), in the presence of plasmonic Au NRs with an average length of 30 nm (4) and in the presence of plasmonic Au NRs with an average length of 35 nm (5) – (b)

3.2. Spectral-luminescent properties

In UV-Vis absorption spectrum of Ag₂S QDs in the region of 680 nm, a clearly expressed feature, corresponding to the ground exciton transition in the optical absorption of QDs, characteristic of charge carriers with confinement in nanocrystals was observed (Fig. 1e, curve 1). UV-Vis absorption spectrum corresponded to Ag₂S QDs with an average size of 2.7 nm [28], which was in good agreement with the TEM image data (Fig. 1a). The studied colloidal Ag₂S QDs were characterized by luminescence with a peak at 820 nm (Fig. 1e, curve 2). The Stokes shift was 0.31 eV (140 nm). According to the data of study [29], the luminescence of Ag₂S QDs is the result of radiative hole recombination with electrons localized at the levels of structural impurity defects.

The morphology and average length of Au NRs of 30, 35, and 45 nm provided the location of longitudinal plasmon resonance peaks at 710, 740, and 825 nm, respectively (Fig. 1e, curves *3*, *4*, *5*). Thus, Au NRs with an average length of 30 and 35 nm provided a detuning of the spectral resonance of the longitudinal plasmon peak of Au NRs from the luminescence band of Ag₂S QDs at

110 and 80 nm, respectively (Fig. 1e, curves 2, 3, 4). The Au NRs with a length of 45 nm provided significant spectral resonance of the longitudinal mode peak of Au NRs with a luminescence spectrum of Ag_2S QDs (Fig. 1e), curves 2, 5).

The formation of plasmon-exciton structures based on Ag₂S QDs and Au NRs, providing significant spectral overlap of the peak of longitudinal plasmon resonance of NRs and the luminescence band of QDs, led to a decrease in the intensity of the Ag₂S QDs luminescence by 2.5 times (Fig. 2a, curves 1-3) at constant luminescence decay time of Ag₂S QDs (Fig. 2a, inset, curves 1, 2). The observed regularities were usually interpreted as photoinduced charge transfer from Ag₂S QDs to Au NRs [18, 26, 27]. At the same time, the authors of the study [30], for the simple classical model of forced and spontaneous dipole transitions in a two-level system near a metal NPs, showed that a decrease in the of luminescence intensity of emitter while maintaining its lifetime near the surface of the metal NPs can be caused by the high absorption of the NPs wave energy. In this case, an increase in the distance between the components of the plasmon-exciton mixture in some cases leads

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to an increase, and in others to a decrease in the luminescence intensity of emitter, which is determined by the mutual orientation of the dipole moment of the transition of the emitter from the ground to the excited state and the field of the metal NPs, which strongly depends on the distance between the components of the mixture [30]. Spatial separation of mixture components by introduction of a polymer into a plasmon-exciton mixture provides an increase in the quantum yield of Ag₂S QDs luminescence from 5 to 7.5% (Fig. 2a, curves 2, 4). A slight increase in the luminescence quantum yield may be due to a significant contribution of the constant luminescence of Ag₂S QDs to the resulting luminescent properties of plasmon-exciton mixtures, free from interaction with Au NRs. The increase of the quantum yield of Ag₂S QDs luminescence in the presence of plasmonic Au NRs was accompanied by reduction of the luminescence decay time from 77 to 58 ns (Fig. 2a, inset, curves 1, 3), which may also indicate the implementation of the Purcell effect in the studied plasmon-exciton mixtures [12, 13, 18-21, 31].

The most interesting regularities were found in the case of plasmon-exciton mixtures, based on Ag₂S QDs and metallic Au NRs with the length of 30 and 35 nm, providing the spectral resonance shift of the NRs extinction from the QDs luminescence band by 80–110 nm (Fig. 2a, curves 1-3). In this case, an asymmetry of the luminescence contour of Ag, S QDs was observed. This asymmetry was manifested in a decrease in the peak luminescence intensity (820 nm) with a simultaneous increase in intensity at the band edge in the region of 720 nm (Fig. 2a, curve 4) or in the region of 900 nm (Fig. 2a, curve 5) with spectral resonance shift at 110 and 80 nm, respectively. In this case, an increase in the average luminescence decay time from 94 to 115 ns was established at a wavelength of 720 nm (900 nm) (Fig. 2a, inset, curves 1, 2) and decrease in the average luminescence decay time from 94 to 16 ns was revealed at a wavelength of 820 nm (Fig. 2a, inset, curves 1, 3). The observed spectral regularities were a manifestation of exciton-plasmon interaction and can be interpreted as the Fano effect [13, 15–17]. The non-monotonic change of the luminescence spectral contour of Ag₂S QDs when changing the spectral resonance shift of the plasmonic peak of Au NRs from the Ag_2S ODs luminescence band within 30 nm (Fig. 2b, curves 1, 2, 4, 5) can be caused by dispersion in the size and distances between components in the plasmon-exciton mixture. Thus, the obtained experimental data certainly require a theoretical description for each specific case. The determination of the parameters of the emitters ensemble and plasmonic nanoparticles for the theoretical description of the experimentally observed effects of plasmon-exciton interaction is a complex task, which requires separate and detailed consideration.

4. Conclusions

The study presents experimental data demonstrating the manifestation of plasmonexciton interaction effects in the luminescent properties of colloidal Ag₂S QDs depending on the degree of overlap of their luminescence band with the plasmon resonance peak of metallic Au NRs. It was established that, under the condition of spectral resonance of the corresponding peaks, control of the luminescence quantum yield is achieved by changing the distance between components of the plasmon-exciton mixture. The spectral resonance shift by 80–110 nm due to a change in the length of the Au NRs leads to a decrease in the luminescence quantum yield of of Ag₂S QDs and asymmetry of the luminescence contour of Ag₂S QDs. Such luminescent properties are the result of the implementation of the Fano effect, the spectral manifestation of which is complicated by the influence of the dispersion of QDs in the ensemble and mutual distances of the components.

Author contributions

Grevtseva I. G. – problem statement, methodology development, conducting research, writing and editing text. Ovchinnikov O. V. – scientific supervision, research concept, development of methodology, writing of text, final conclusions. Smirnov M. S. – research concept, research implementation, review writing and editing of the text. Aslanov S. V. – conducting research, review writing. Astashkina M. S. – conducting research, editing of the text.

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Conflict of interests

The authors declare that they have no known competing financial interests or personal relationships that could have influenced the work reported in this paper.

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